

Electron-Electron Scattering Effects in the Low-Temperature Lorenz Numbers of Dilute Pd-Ni Alloys

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September 15, 1969



NAVAL RESEARCH LABORATORY
Washington, D.C.

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ABSTRACT

The thermal and electrical resistivities of Pd-Ni alloys at concentrations ranging from 0 to 1.0 at-% Ni have been measured at temperatures between 2 and 20°K. The Lorenz numbers L_e appropriate to electron-electron scattering have been extracted from the data and are found to be relatively insensitive to Ni concentration. A theoretical calculation of L_e has been carried out under the assumption that the entire current is carried by *s* electrons which are scattered from spin density fluctuations of the electrons in the *d* band. A consistent interpretation of the data is obtained by employing the local enhancement model, while the uniform enhancement model is found to yield values of L_e which decrease too rapidly with increasing Ni concentration to reasonably represent the experimental results. The calculated value of L_e is significantly smaller than the experimental value for pure Pd; it is felt this may be due to the neglect of detailed band structure effects in this work. Lorenz numbers for Baber scattering have also been calculated by using Coulomb interaction screened by the *d* electrons. It is found that the presence of Baber scattering cannot remove the discrepancy between theory and experiment for pure Pd. The Baber scattering model is also used to calculate the L_e for rhenium and gives very good agreement with experiment.

PROBLEM STATUS

This report completes one phase of the problem; work on other aspects of the problem is continuing.

AUTHORIZATION

NRL Problem No. M01-10
Project RR 007-01-46-5408

Manuscript submitted June 17, 1969.

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INTRODUCTION

It has been known for some time that the T^2 dependence of the electrical resistivity at low temperatures in nonmagnetic transition metals can be interpreted as arising from the scattering of those electrons primarily responsible for carrying the current by electrons which exhibit much larger effective mass and which lie in other bands. The first theoretical calculation of this scattering process was Baber's (1) analysis of de Haas and de Boer's observation of a T^2 term in the electrical resistivity of platinum. Baber considered an s band of conduction electrons scattered by a screened Coulomb interaction with the much heavier d-band electrons which make only a small contribution to the transport current. Although the T^2 term in the electrical resistivity of many transition metals has been observed (2) since this early work, it was not until very recently that the corresponding contribution to the thermal resistivity, a term linear in T , was experimentally established (3-7). With the observation of the electron-electron thermal resistivities, it becomes feasible to study electron-electron scattering in the appropriate metals by use of the Lorenz number; i.e., the ratio of the electrical resistivity to the product of the thermal resistivity with the absolute temperature. This provides a useful characterization of the scattering, for the Lorenz number is sensitive to the angular distribution of the scattering, while certain quantities whose magnitudes are difficult to estimate on theoretical grounds drop out of the ratio. Furthermore, by making experimental observations of both the thermal and the electrical resistivity of the same specimen and with the same probes, the difficult task of accurately determining specimen geometries is circumvented.

The dilute Pd-Ni alloy system appears to be a very appropriate one for a study of electron-electron scattering effects employing Lorenz number measurements because, as shown by Schindler and Rice (8), the electron-electron term in the electrical resistivity of palladium is strongly enhanced by the addition of small amounts of nickel. In their original presentation of the data, Schindler and Rice used a model in which the current-carrying s electrons were scattered by spin density fluctuations of the electrons in the d band. They postulated that the effect of the nickel could be represented by increasing the exchange enhancement in the entire matrix in some average fashion; i.e., they employed the so-called "uniform enhancement" model. Lederer and Mills (9) later showed that the data could also be interpreted in terms of the scattering of s electrons from spin density fluctuations by assuming that enhancement due to the addition of nickel occurred in the vicinity of the nickel impurity. In fact, they showed that this "local enhancement" model gave better correlations between the electrical resistivity and magnetic susceptibility data than had been obtained from the uniform model. We have undertaken a study of the Lorenz numbers of these alloys in the hope of obtaining more information concerning the relevance of these models to the effect of electron-electron interactions on the transport properties of this (and similar) systems.

From an experimental point of view, the observation of the electron-electron term in the thermal resistivity of the alloys is expected to be complicated by the possible presence of a significant amount of heat transport via the lattice. The work of Schindler and Rice indicates, however, that in the dilute Pd-Ni alloys the electron-electron term would be expected to be sufficiently large that it would still be discernible in the presence

of a lattice heat conduction of typical size for transition metal alloys. In other, less-enhanced, alloy systems, the detailed behavior of the electron-electron term would be expected to be masked by the phonon term. Moreover, since the determination of the Lorenz number L_e for electron-electron scattering is particularly well suited to a differentiation between the uniform and local models, as we shall see below, it was felt that the L_e values of these alloys would be of considerable interest.

This report is presented in five sections. In the next (second) section, we employ a simple two-band model to calculate expressions for L_e for Pd-Ni at low temperatures for both uniform and local enhancement. Although terse presentations of results of similar calculations have already appeared (8,10-13), we present here a discussion in some detail because this type of calculation is central to a study of the effects of electron-electron interaction on the Lorenz number. Furthermore, a full discussion of this calculation is appropriate in that, for reasons which shall be presented, we find ourselves in disagreement with features of the earlier work. In the third section, we present the results of measurements of the thermal and electrical resistivities between 2 and 20°K of Pd-Ni specimens ranging from pure Pd to Pd-1.0 at-% Ni. The fourth section contains a development of specific predictions for the L_e of Pd-Ni from the theory of the second section. We find that the local enhancement model provides a consistent interpretation of the alloy data, whereas the uniform enhancement model predicts a more rapid fall of L_e with increasing Ni concentration than is experimentally observed. We also found that the L_e of pure Pd is not well estimated by our simple model of scattering from spin density fluctuations. Here we also calculate L_e for the case of electrons interacting via a simple screened Coulomb interaction, that is when the scattering is the so-called Baber scattering, and we find that Baber scattering will not resolve the discrepancy between theory and experiment observed in pure Pd. In the last section, we consider some of the implications of this work with regard to other types of measurements and other transition metals. In particular, our model of Baber scattering is found to yield an estimate of L_e for rhenium which fits well with the experimental value.

THEORETICAL CONSIDERATIONS

In this section we examine the theory of the electron transport coefficients of transition metal alloys, with application to the theory of the Lorenz number in mind. We shall employ the simple *s-d* model in this discussion (14). That is, one imagines that the Fermi surface of the material has an *s*-like portion, nearly free-electron in character, and also a *d*-like portion. Since the *d* band is much narrower than the *s* band, the effective mass of the *d* electrons is large compared to that of the *s* electrons. Thus, the transport currents are carried primarily by the *s* electrons. Since the contribution to the density of states at the Fermi level from *d* electrons is large compared to that of the *s* electrons, the magnetic properties of the material as well as the specific heat are determined primarily by the *d* electrons. The *s-d* model has been widely used to interpret the observed properties of transition metals and their alloys (1,8-15). The model is of dubious validity for host metals in the middle of the transition metal series, since *s-d* hybridization is important in this portion of the periodic table. However, for the elements near the right end of the periodic table, band-structure calculations (16) indicate that this simple model provides a reasonable qualitative picture of the nature of the Fermi surface, although the *d*-like portion is apparently very complex in shape.

A number of authors have considered the contributions to the electrical and thermal resistivities of transition metals from electron-electron interactions (1,8-15). We denote the contributions to the electrical and thermal resistivity from electron-electron scattering by ρ_e and w_e , respectively. In most of this work, two mechanisms have been considered as providing the principal contributions to ρ_e and w_e . Baber (1) considered the scattering of an *s* electron by *d* electrons resulting from the direct Coulomb

interaction between the particles. This mechanism has recently been reexamined by Rice (11). One may regard this scattering process as resulting from the interaction of the s electron with the number density fluctuations in the d band. Recently, it has been pointed out (8-10,15) that in materials which are strongly exchange enhanced, the scattering of s electrons from the large-amplitude spin density fluctuations in the d band can dominate ρ_e and w_e .

In this section, we construct a theory of the Lorenz number $L_e = \rho_e / (w_e T)$ of Pd and Pd-Ni alloys, assuming that the principal contribution to ρ_e and w_e comes from the spin density fluctuations in these strongly exchange-enhanced materials. Two models have been introduced to explain the transport coefficients of this alloy system. The uniform enhancement model assumes that the effect of adding Ni may be represented by a change in the exchange enhancement factor of the matrix (8). The local enhancement model treats the alloy in a scheme valid in the dilute limit, where the spatial inhomogeneity of the spin density fluctuations in the vicinity of the Ni site is taken into account. It has been pointed out (9) that the local enhancement model appears to provide a more consistent interpretation of the magnitude of the effect of alloying on ρ_e , and also of the relationship between the concentration dependence of the coefficient A of the T^2 term in the electrical resistivity and that of the magnetic susceptibility χ . In this section, we shall present calculations of the concentration dependence of L_e for both models. We find that the local enhancement model offers a reasonable value for the Lorenz number of the alloys in the concentration region where the spin fluctuation contribution to ρ_e and w_e is dominated by scattering in the vicinity of the Ni sites. However, we find that in pure Pd, the spin fluctuation scattering leads to a value of L_e that is smaller than the observed value. The uniform enhancement model leads to a value for L_e that decreases rapidly with increasing Ni concentration, in disagreement with the data. Of course, the two models lead to the same value of L_e for the pure matrix.

We also examine the contribution to the Lorenz number from Baber scattering in the pure matrix. We find that if Baber scattering dominates ρ_e and w_e in the pure metal, the predicted L_e is still significantly smaller than the observed value. This discrepancy may be resolved if band-structure effects are properly taken into account. Rice (11) has examined the contribution to L_e from Baber scattering, assuming the s - d Coulomb interaction is statically screened by the s electrons. However, in materials with partially filled d bands, the static dielectric constant $\epsilon(q, 0)$ is dominated by the d band (17), since the Fermi-Thomas screening wave vector κ associated with a given band is proportional to the contribution from that band to the density of states at the Fermi level. When the s - d interaction is screened by the d electrons, the value of L_e that obtains differs from the values given by Rice (11). Also, in these metals the coefficient A of the T^2 term in ρ_e is not proportional to the square of the density of states at the Fermi level (11), since an increase in the density of states shortens the Fermi-Thomas screening length and decreases the value of the effective s - d matrix element.

Before we proceed to the quantitative discussion, we present some simple, intuitive considerations that will illustrate the effect of electron-electron scattering on the Lorenz number. This will provide a useful reference when we examine the effect of various factors that enter the quantitative theory of L_e .

For this purpose, we apply the diagrammatic scheme employed by Klemens (18) in his discussion of the transport properties of metals. In the steady state, the distribution of the conduction electrons in k space under the influence of either a thermal gradient or an electric field can be visualized crudely as shown in Fig. 1, where we assume a spherical Fermi surface for simplicity. We let the solid dots represent an increase in the number of electrons over the "zero field" value, and the open circles indicate a decrease in this number. This type of crude picture permits a qualitative understanding of the effects of various kinds of scattering on the Lorenz number.

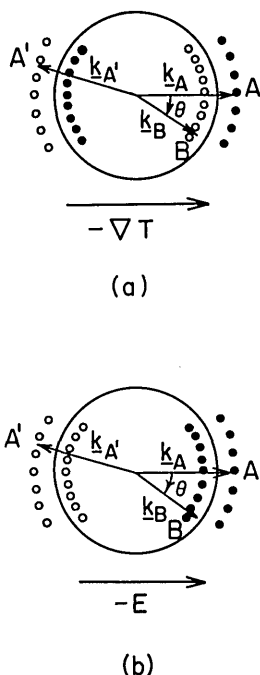


Fig. 1 - Schematic representation of electron distributions in the steady state under the assumption of a spherical Fermi surface. The case of a thermal gradient is depicted in part a, while that of an electric field is illustrated in part b. Solid dots represent an excess number of electrons over the distribution in the absence of external forces, whereas open circles represent a deficiency with respect to this distribution. Both dots and circles are intended to be within a thermal layer of the Fermi surface.

Consider first the case of elastic scattering. An electron in state A can relax only to states on the same constant energy surface, if only elastic scattering is present. A typical transition of this type would be a scattering from the initial state A to a final state A', as indicated in Fig. 1. Klemens refers to this kind of transition as a "horizontal" transition. As long as the linearized Boltzmann equation can be used to describe the transport process, it is clear that the thermal conductivity relaxation time and the electrical resistivity relaxation time are the same for the case of elastic scattering. This is because the time required for the relaxation to equilibrium of the states on a given constant energy surface is independent of the amplitude of the deviation from equilibrium on this constant-energy surface. As indicated in Fig. 1a and 1b, the angular variation of the change in distribution of particles on any particular constant-energy surface is the same for both the electrical resistivity and the thermal resistivity. It is only the amplitudes that differ. Thus, as long as the energy dependence of the relaxation time may be ignored, the electrical and thermal relaxation times must be the same, and the classical Sommerfeld value L_s of the Lorenz number is realized.

Next consider the Lorenz number L_e from electron-electron scattering. A given electron can suffer energy changes in a collision with another electron. We will be interested in the case where a current-carrying s electron collides with an electron in the partially filled d band. This allows s-electron transitions of the type A → B in Fig. 1a. One can see that the thermal conductivity and the electrical conductivity relaxation times are now different. An extreme example of this difference is the well-known case of the contribution to ρ and w from electron-phonon collisions at low temperatures, where only long-wavelength phonons are thermally excited. In this regime, the electron distribution is relaxed predominantly by small-angle scattering events. The effective relaxation time for the electrical resistivity ρ becomes longer than that for the electronic contribution to w by a factor of roughly $(\theta_D/T)^2$, producing a Lorenz number that vanishes as $T \rightarrow 0$. By analogy with the scattering of electrons by phonons, we may expect the Lorenz number L_e defined above to be reduced from the Sommerfeld value L_s ,

because of the inelastic nature of the electron-electron collisions.* However, there are important differences between the two cases, primarily because in electron-electron scattering, large-angle scatterings contribute to the transport relaxation time even at low temperatures.

Let us consider the qualitative features of the electron-electron scattering. Suppose we examine the contribution to the transport current from an *s* electron in the state A of Fig. 1. For simplicity, let the wave vector \mathbf{k}_A of this state be directed parallel to the transport current. Suppose the electron is scattered by a *d* electron through an angle θ to some final state B, as indicated in Fig. 1. If the collision is inelastic, the final-state energy E_B does not equal the initial energy E_A . For a spherical Fermi surface, this collision changes the contribution of state A to the electrical current j_ρ by the amount (with $\hbar = 1$)

$$\Delta j_\rho = \frac{e}{m^*} \hat{x} \cdot (\mathbf{k}_A - \mathbf{k}_B) \approx \frac{ek_F}{m^*} (1 - \cos \theta) . \quad (1)$$

In Eq. (1) m^* denotes the effective mass, e the charge, and k_F the Fermi wave vector of the current-carrying electrons, while \hat{x} is a unit vector along the direction of the current. Now consider the effect of the same collision on the energy transport current j_w . Noting that the contribution of a given state to the heat current (19) is $(E - \mu) k/m^*$, where μ is the Fermi energy, we have

$$\begin{aligned} \Delta j_w &= \frac{\hat{x}}{m^*} \cdot [\mathbf{k}_A(E_A - \mu) - \mathbf{k}_B(E_B - \mu)] , \\ &= \frac{\hat{x}}{m^*} \cdot [(\mathbf{k}_A - \mathbf{k}_B)(E_A - \mu) + \mathbf{k}_B(E_A - E_B)] , \\ &\approx \frac{k_F}{m^*} [(1 - \cos \theta)(E_A - \mu) + \cos \theta(E_A - E_B)] . \end{aligned} \quad (2)$$

Both terms in this last expression are of the same order of magnitude for the most important values of E_A and E_B . We may think of the collision as a two-step process. First, the electron is scattered through the angle θ , with no change in energy. This alters the heat current by the amount indicated in the first term of Eq. (2). Then the electron changes its energy by the amount $(E_A - E_B)$. This gives the second contribution to Δj_w indicated in Eq. (2). In the case of the electrical resistivity, only the first step is effective in changing the transport current, since the small shift in velocity associated with the second "jump" can be neglected to a good approximation. Thus, crudely speaking, a given inelastic collision of the type described is twice as effective in reducing the heat current as it is in reducing the electrical current. As a consequence, the Lorenz number L_e defined above should be roughly half of the classical Sommerfeld value L_s . Of course, for elastic scattering, the second term of Eq. (2) is missing, and a given collision is equally effective in reducing both transport currents. One then obtains the classical Lorenz number L_s as a consequence.

Any feature of the scattering matrix element which peaks the scattering in the forward direction will decrease the quantity L_e below the value $\approx L_s/2$ suggested by the above argument, since this enhances the relative contribution of the "second step" (the vertical step in Klemens's language) to the thermal resistivity relaxation rate. In the

*Herring has argued that for transition metals with Fermi surfaces of complex geometry, with all portions of the Fermi surface contributing to the transport currents, one should find $(L_e/L_s) \approx 2/3$ in the absence of impurity scattering. (See Ref. 9.) We shall comment on the applicability of Herring's argument to the present system later in this section.

case of phonon scattering at temperatures low compared to the Debye temperature θ_D , one has an extreme example of this effect, since only scattering through angles less than $\theta_c \approx (T/\theta_D) \ll 1$ is important. It is easy to see that the second term of Eq. (2) dominates Δj_w , so that one obtains the well-known result

$$\frac{L_{\text{phonon}}}{L_s} \approx \frac{\int_0^{\theta_c} (1 - \cos \theta) \sin \theta d\theta}{\int_0^{\theta_c} \sin \theta d\theta} = \frac{1}{4} \theta_c^2 \approx \left(\frac{T}{\theta_D}\right)^2$$

For the case of phonon scattering, the temperature dependence of L_{phonon} arises because of the temperature dependence of the cutoff angle θ_c . For electron-electron scattering, the matrix element is temperature independent to a good approximation, and as a consequence, the Lorenz number L_e is temperature independent.

We now turn to the detailed calculation of w_e and ρ_e . As we mentioned, we first assume that in the strongly exchange-enhanced Pd-Ni system, the dominant contribution to ρ_e and w_e comes from the scattering of *s* electrons by the large-amplitude, spin-density fluctuations in the *d* band (8,9,15). Since the details of the calculation of ρ_e have appeared in the literature, we present a detailed discussion of only the computation of the thermal resistivity w_e .

A typical scattering event is indicated in Fig. 2. This diagram illustrates spin-flip scattering of an *s* electron from the exchange-enhanced transverse fluctuations in spin density in the *d* band. The wavy line is the *s-d* exchange interaction J (15), and the dotted lines indicate the interaction between the excited particle and the hole in the *d* band. Of course, in the theory of the transport coefficients, nonspin-flip scattering from longitudinal fluctuations must also be included. We shall assume that only the *s* electrons carry the currents, as mentioned above. Consequently the *d* electrons will be treated as if they were in thermal equilibrium.

We shall employ the variational method discussed by Ziman, and write the electron-electron contribution to w_e in the form (20)

$$w_e = \frac{1}{2k_B} \frac{\sum_{k\sigma} \sum_{k'\sigma'} [\Phi(k\sigma) - \Phi(k'\sigma')]^2 P(k\sigma \rightarrow k'\sigma')}{\left| \sum_{k\sigma} \mathbf{V}(k\sigma) [E(k\sigma) - \mu] \Phi(k\sigma) \frac{\partial f^0(k\sigma)}{\partial E(k\sigma)} \right|^2}. \quad (3)$$

The quantity k_B is Boltzmann's constant, and σ is a spin index. In Eq. (3),

$$P(k\sigma \rightarrow k'\sigma')$$

is the number of transitions per unit time from state $(k\sigma)$ to $(k'\sigma')$ when the system is in thermal equilibrium, i.e.,

$$P(k\sigma \rightarrow k'\sigma') = [1 - f^0(k'\sigma')] f^0(k\sigma) W(k\sigma \rightarrow k'\sigma'),$$

where $W(k\sigma \rightarrow k'\sigma')$ is the transition rate for the case when $(k\sigma)$ is occupied and $(k'\sigma')$ is empty, and $f^0(k\sigma)$ is the equilibrium occupation number. The deviation of the distribution function from equilibrium is written

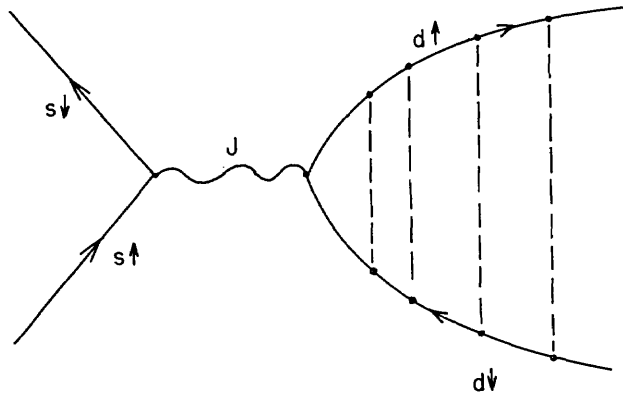


Fig. 2 - Diagram of a typical spin-flip scattering of an s electron from the exchange-enhanced transverse fluctuation in spin density in the d band. The wavy line represents the s - d exchange interaction J (15), and the dotted lines indicate the interaction between the excited particle and hole in the d band.

$$f(k\sigma) = f^0(k\sigma) - \Phi(k\sigma) \frac{\partial f^0(k\sigma)}{\partial E(k\sigma)}.$$

In the denominator of Eq. (3), $V(k\sigma) = \nabla_k E(k\sigma)$, and μ is the chemical potential.

Since we shall consider only paramagnetic alloys in zero magnetic field, $\Phi(k\sigma)$ and $f^0(k\sigma)$ are independent of spin. We then define

$$P(k \rightarrow k') = \sum_{\sigma\sigma'} P(k\sigma \rightarrow k'\sigma')$$

and write the expression for w_e in the form

$$w_e = \frac{1}{8k_B} \frac{\sum_{kk'} [\Phi(k) - \Phi(k')]^2 P(k \rightarrow k')}{\left| \sum_k V(k) [E(k) - \mu] \Phi(k) \frac{\partial f^0(k)}{\partial E(k)} \right|^2}. \quad (4)$$

To compute w_e , we take for the function $\Phi(k)$ an expression of the form

$$\Phi(k) = \mathbf{u} \cdot \mathbf{V}(k) [E(k) - \mu], \quad (5)$$

where \mathbf{u} is a vector directed parallel to the heat current. We then evaluate Eq. (4) by assuming the s -electron energy to be that appropriate for a spherically symmetric, parabolic band.

Before proceeding with the details of evaluating the expression in Eq. (4), some brief comments are in order. Equation (4) may be derived by computing the rate at which entropy is produced by the collisions of the s electrons. In addition to the electron-electron scattering under consideration in this section, impurity scattering is also

present. In fact, the electron-electron contribution to w is small compared to the contribution from impurity scattering, for the temperature range of interest in this work.

If for $P(k \rightarrow k')$ we were to take the sum of the impurity scattering rate and the scattering rate from the spin fluctuations in the d band, and if for $\Phi(k)$ we were to insert the exact solution to the Boltzmann equation, we of course would obtain an exact expression for the total thermal resistivity w from Eq. (4). In the presence of impurity scattering only, one knows the linearized Boltzmann equation can be solved exactly if the constant-energy surfaces are spherical (20). The solution has the form of Eq. (5) if the energy dependence of the relaxation time can be ignored. When the electron-electron scattering rate is small compared to the impurity scattering rate, one may compute the change in the rate of entropy production to first order in the electron-electron scattering rate by using perturbation theory. One obtains the expression in Eq. (4), where $\Phi(k)$ is the exact form for a spherical Fermi surface with impurity scattering only, and $P(k \rightarrow k')$ is the scattering rate from electron-electron collisions. The structure of the theory is analogous to the Schrödinger equation, with the entropy analogous to the energy, and the scattering kernel to the Hamiltonian. The change in energy to first order in some small perturbation is found by averaging the perturbation over the unperturbed wave function. These remarks also apply to the calculation of the electron-phonon scattering contribution to w , provided this contribution is small compared with the impurity scattering background. This is consistent with the manner in which we separate the measured total resistivities into their component parts.

The denominator in Eq. (4) is readily evaluated. One finds

$$\begin{aligned} \mathcal{D} &= 4 \left| \sum_{\mathbf{k}} \mathbf{V}(\mathbf{k}) [E(\mathbf{k}) - \mu] \Phi(\mathbf{k}) \frac{\partial f^0(\mathbf{k})}{\partial E(\mathbf{k})} \right|^2, \\ &= u^2 \frac{k_s^6}{81 m_s^2} (k_B T)^4, \end{aligned} \quad (6)$$

where k_s is the Fermi wave vector of the s band, and m_s is the effective mass of the s electrons.

Consider the numerator next. For the paramagnetic metal,

$$\sum_{\sigma\sigma'} W(k\sigma \rightarrow k'\sigma') = 2W(k\uparrow \rightarrow k'\uparrow) + W(k\uparrow \rightarrow k'\downarrow) + W(k\downarrow \rightarrow k'\uparrow).$$

For the model under consideration, one finds (15)

$$\begin{aligned} W(k\uparrow \rightarrow k'\downarrow) &= \frac{J^2}{4N} \int_{-\infty}^{\infty} dt e^{-i\Omega(k,k')t} \langle S_{(-)}(k' - k, 0) S_{(+)}(k' - k, t) \rangle_T, \\ W(k\downarrow \rightarrow k'\uparrow) &= \frac{J^2}{4N} \int_{-\infty}^{\infty} dt e^{-i\Omega(k,k')t} \langle S_{(+)}(k - k', 0) S_{(-)}(k - k', t) \rangle_T. \end{aligned}$$

In these expressions, J is the magnitude of the s - d exchange interaction, N the number of unit cells in the crystal, $\Omega(k, k') = E(k) - E(k')$, and $S_{(+, -)}(q, t)$ is the operator that describes the appropriate transverse component of spin density in the d band of wave

vector q . The scattering rate from longitudinal fluctuations is easily derived, using the previous techniques:

$$W(k\uparrow \rightarrow k'\uparrow) = \frac{J^2}{4N} \int_{-\infty}^{\infty} dt e^{-i\Omega(k,k')t} \langle S_z(k-k', 0)^\dagger S_z(k-k', t) \rangle_T .$$

From detailed balance considerations, one has (15)

$$W(k\downarrow \rightarrow k'\uparrow) = e^{\beta[E(k)-E(k')]} W(k'\uparrow \rightarrow k\downarrow) ,$$

where $\beta = 1/k_B T$.

Since the Hamiltonian is invariant under spin rotations, and since the ground state is presumed to be paramagnetic, one easily sees that

$$W(k\uparrow \rightarrow k'\uparrow) = \frac{1}{4} [W(k\downarrow \rightarrow k'\uparrow) + W(k\uparrow \rightarrow k'\downarrow)] .$$

The expression for the thermal resistivity w_e may then be simplified to the form

$$w_e = \frac{3}{2k_B T} \sum_{kk'} [\Phi(k) - \Phi(k')]^2 f^0(k) [1 - f^0(k')] W(k\uparrow \rightarrow k'\downarrow) .$$

We now employ the specific form of $W(k\uparrow \rightarrow k'\downarrow)$. If the Bloch functions of the d electrons may be treated in the tight binding approximation, then we may write

$$W(k\uparrow \rightarrow k'\downarrow) = \frac{J^2}{4N} \{1 + n[\Omega(k,k')]\} A[(k' - k, \Omega(k,k'))] |F(k' - k)|^2 , \quad (7)$$

where $A(q, \Omega)$ is the spectral density function deduced from the wave vector and the frequency-dependent transverse susceptibility. Explicitly (15),

$$A(q, \Omega) = \frac{1}{i} [\chi(q, \Omega - i\epsilon) - \chi(q, \Omega + i\epsilon)] ,$$

where ϵ is a positive, infinitesimal quantity. The quantity $F(q)$ is the form factor of the Wannier function $\Phi(r)$ associated with the unit cell centered at the origin; that is,

$$F(q) = \int d^3r \exp(iq \cdot r) |\Phi(r)|^2 .$$

In Eq. (7), $n[\Omega(k,k')]$ is the Bose-Einstein factor, evaluated at the frequency $\Omega(k,k')$.

In the present calculation, which considers only the low-temperature form of the transport coefficients, we will encounter collisions with energy transfers of the order of $k_B T$, where T is at most 20°K. Thus, we may replace $A(q, \Omega)$ in Eq. (7) by its low-frequency limiting form. Since it is well known that $A(q, \Omega)$ is an odd function of frequency, one can write the low-frequency form as

$$A(q, \Omega) = \Omega \alpha(q) . \quad (8)$$

We then evaluate w_e by employing Eq. (7) and by using the low-frequency form of $A(q, \Omega)$ given in Eq. (8). One more simplification is convenient. The form factor $F(q)$ will depend in general on both the magnitude and the direction of q , if we choose $F(q)$ to be the form factor of a realistic d orbital. We shall assume that $F(q)$ depends only on

$|\mathbf{q}|$ and ignore the dependence on the direction of \mathbf{q} . To include the dependence of F on the direction and magnitude of \mathbf{q} would greatly complicate the numerical computations to be presented later. We make the same simplification for the function $\alpha(\mathbf{q})$ defined in Eq. (8).

With these approximations and the assumption that the s band is parabolic, one may evaluate w_e in a straightforward fashion. We find

$$w_e = \frac{81 J^2 T}{64 \pi^4 \nu \hbar v_s^2} [I_b \beta(3) + I_a \beta(1)] , \quad (9)$$

where

ν is the number of unit cells per unit volume,

ν_s is the Fermi velocity of the s electrons,

and

$$I_a = \int_{-\infty}^{\infty} dx dx' (x - x')^3 f^0(x) [1 - f^0(x')] [1 + n(x - x')] ,$$

$$I_b = \int_{-\infty}^{\infty} dx dx' x x' (x - x') f^0(x) [1 - f^0(x')] [1 + n(x - x')] ,$$

$$\beta(m) = \int_0^{2Q} d\eta \eta^m \alpha(k_s \eta) |F(k_s \eta)|^2$$

Notice that $\beta(m)$ has the dimensions of $1/(\text{energy})^2$. We have introduced the variable $\eta = \sqrt{2} \sqrt{1 - \cos \theta}$, where θ is the angle through which the s electron is scattered. Q is determined by the momentum of the d electrons and is related to the maximum angle, θ_M , through which the s electrons are scattered, by $2Q = \sqrt{2} \sqrt{1 - \cos \theta_M}$. In the case of a spherical d band, $Q = \max \{(k_d/k_s), 1\}$.

The integrals I_a and I_b , together with I_c which appears below, are evaluated in the appendix. We find

$$I_a = 8\pi^4/15 \quad \text{and} \quad I_b = 2\pi^4/15 .$$

One can compute the electrical resistivity ρ_e in the same fashion. We find

$$\rho_e = \frac{9J^2}{64} \frac{(k_B T)^2}{\nu \hbar e^2 v_s^2} I_c \beta(3) , \quad (10)$$

where $\beta(3)$ is defined above, and

$$I_c = \int_{-\infty}^{\infty} dx dx' (x - x') f^0(x) [1 - f^0(x')] [1 + n(x - x')] = 2\pi^2/3 .$$

We now form the expression for the Lorenz number L_e . We have

$$L_e = L_s \left[\frac{5}{3 + 12 \frac{\beta(1)}{\beta(3)}} \right], \quad (11)$$

where L_s is the classical Sommerfeld value of the Lorenz number. Since Eq. (11) involves only the ratio $\{\beta(1)/[\beta(3)]\}$, the Lorenz number L_e depends only on the angular distribution of the scattering.

Two comments should be made concerning the form of Eq. (11). We have argued above that when impurity scattering is strong compared to electron-electron scattering, the procedure we employ gives an exact expression for w_e and ρ_e . Thus, Eq. (11) is also exact, for the simple s - d model, in the limit of strong impurity scattering. Recently, exact solutions of the Boltzmann equation have been exhibited for the case where only electron-electron scattering is present (21). It has been pointed out (11) that in this case, the exact solution shows the Lorenz number L_e is larger than that given by Eq. (11) by a factor of 6/5. However, this theory cannot be used to describe the properties of transition metals at low temperatures, where the electron-electron scattering makes only a small contribution to the total resistivity for materials available so far.

Rice has also examined the theory of the Lorenz number L_e for the scattering of s electrons from d -electron spin fluctuations, when the spin fluctuations are described by the uniform enhancement model (10). Rice treats the transverse spin fluctuations in a manner similar to that of the present work. However, he treats the nonspin-flip scattering that results from the $s_z S_z$ of the s - d exchange interaction as if this scattering were Baber scattering (1). Rice thus obtains an expression for L_e which differs from that in Eq. (11). However, we have seen that the $s_z S_z$ scattering rate may be related to the scattering from transverse fluctuations by simple considerations of rotational invariance, if the metal is paramagnetic and if no external field is present. The effect of including the $s_z S_z$ scattering may be accounted for by simply multiplying the spin-flip scattering rate by 3/2 in both ρ_e and w_e , as we have done above. The factor of 3/2 is well known, and was first pointed out in a slightly different context by Penn (22).

We remark finally that Eq. (11) may be applied to both the uniform enhancement model and the local enhancement model. The two differ by the form of $\alpha(k_s \eta)$, i.e., by $\beta(1)/[\beta(3)]$. This statement assumes that the principal effect of the Ni atom is to change the values of the d - d Coulomb interaction in the impurity cell. In this approximation, one ignores the effect of the change in J and the change in form factor on the transport properties.

EXPERIMENTAL RESULTS

We present in this section the results of thermal and electrical resistivity measurements between 2 and 20°K on Pd-Ni specimens containing approximately 1/4, 1/2, and 1 at-% Ni. The data on pure Pd, which is included here for reference, has appeared in earlier papers (5,6), but was obtained under the same experimental conditions in the same cryostat as the data for the Pd-Ni specimens.

The cryostat employed for these measurements has been described in detail elsewhere (4). Temperature differences were measured with a Au-0.02 at-% Fe vs Ag-0.37 at-% Au thermocouple which was calibrated in a separate experiment with a germanium resistance thermometer. This germanium thermometer was calibrated at the National Bureau of Standards in accordance with the 1965 Provisional Temperature Scale. Electrical resistivities were determined with a four-probe potentiometric system. The

uncertainties in the electrical resistivities are approximately $\pm 0.2\%$, while the thermal resistivities have estimated uncertainties of the order of $\pm 1/2$ percent.* Both electrical and thermal resistivities were measured on the same specimens and with the same contacts, so that the Lorenz number values obtained are essentially free of errors due to inaccuracies in measuring specimen geometries.

The alloys were prepared from Johnson-Matthey palladium sponge and Johnson-Matthey nickel rods of five 9's nominal purity by induction melting in quartz crucibles under an argon atmosphere which was purified by use of a dry-ice and acetone trap. The resultant ingots were then swaged to the desired geometry, rods 6 in. long and roughly $1/8$ in. in diameter. Strain-relief anneals were performed at intermediate steps in the swaging, and a final anneal of 4-1/2 hr at 900°C in a vacuum of about 1×10^{-5} Torr was performed just prior to mounting the specimens in the cryostat.

X-ray fluorescence analysis of the completed specimens indicated that the nickel concentrations of each rod were uniform to within ± 0.1 at-%. A more accurate wet chemical analysis of both ends of each specimen was also performed, and the results indicated an end-to-end compositional uniformity of within ± 0.02 at-%. A wet chemical analysis of the iron impurity content of each sample was also made. The results indicated that the Fe impurity content appears to increase with increasing Ni concentration. A tabulation of the wet analysis results appears below.

Nominal Composition (at-% Ni)	Left-end Composition (at-% Ni)	Right-end Composition (at-% Ni)	Fe Content (ppm by wt)
0.21	0.20	0.23	2
0.5	0.47	0.48	2
1.0	0.96	1.00	5

In analyzing the measured resistivities, we shall make use of Matthiessen's rule in a general sense and separate the total resistivities into the sums of terms arising from the scattering of the electrons by impurities, phonons, and other electrons. As discussed in the second section, this separation is appropriate when the impurity scattering is much larger than the scattering by the electrons or the phonons. Thus we shall extract values of the electron-electron terms for comparison with the above theory by concentrating on the data at the lower temperatures, where the impurity scattering is dominant.

We have, therefore, compared the measured electrical resistivities (ρ) at the lower temperatures to an equation of the form

$$\rho = \rho_0 + AT^2 + BT^5, \quad (12)$$

where ρ_0 is the impurity resistivity, T the absolute temperature, AT^2 the dominant electron-electron scattering term, and BT^5 the result of the scattering of electrons by phonons. The results are presented in Fig. 3 in a plot of $(\rho - \rho_0)/T^2$ vs T^3 . These results are in agreement with those of Schindler and Rice (8), in that we observe A values which increase and B values which decrease with increasing Ni concentration, as shown in Table 1. The values of A , B , and ρ_0 have been obtained from least-squares fits of the data at the lower temperatures to Eq. (12).

*As one of the authors (J. T. Schriempff) has pointed out (Ref. 6), the present stage of development of standards for the absolute temperature scale limits the accuracy of thermal resistivity determinations to approximately 99-1/2%.

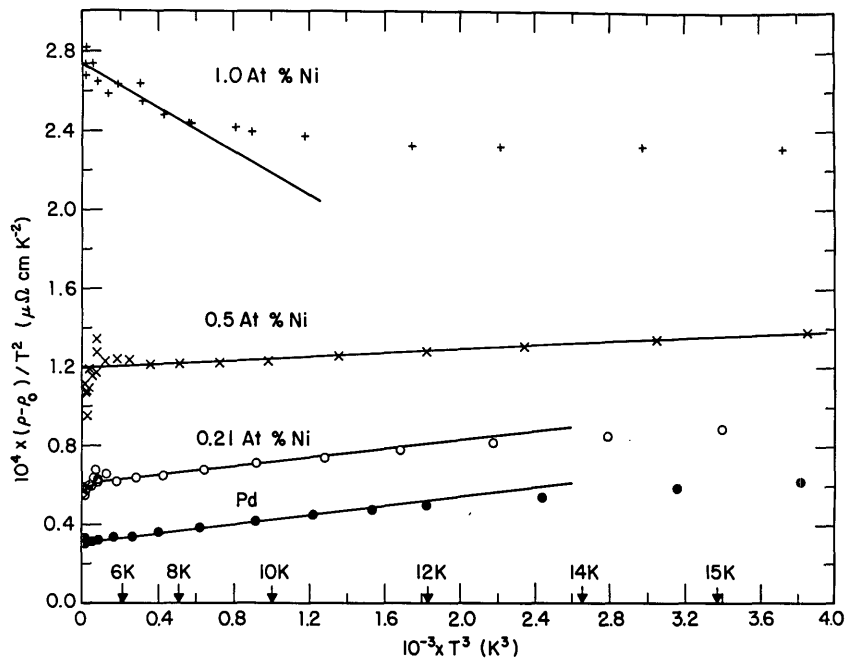


Fig. 3 - Electrical resistivity ρ of the Pd-Ni alloys (with Ni concentrations as indicated) as a function of the cube of the absolute temperature T . The straight lines were obtained by a least-squares comparison of the data at the lower temperatures to the equation $\rho = \rho_0 + AT^2 + BT^5$.

Turning now to the thermal resistivity, if we again assume that resistivities due to various scattering mechanisms are additive, the thermal resistivity w at low temperatures can be represented by

$$w = \frac{C}{T} + \alpha T + \beta T^2. \quad (13)$$

In Eq. (13), C/T is the impurity scattering term, αT the electron-electron scattering term, and βT^2 represents the effect of scattering of electrons by phonons. Ignoring the possibility of phonon conduction for the moment, the data can be compared to Eq. (13) by plotting $(wT - C)/T^2$ vs T . This has been done in Fig. 4, where values of C have been obtained by simply extrapolating the data to 0°K in a plot of wT vs T^2 . These C values yield Lorenz numbers (L_0) in the $T = 0$ limit which are shown in Table 1 and are in reasonable accord with the Sommerfeld value of $2.443 \times 10^{-8} \text{ V}^2/^\circ\text{K}^2$. The existence of an electron-electron term, as evidenced by a nonzero value of $(wT - C)/T^2$ as T approaches zero, is clearly evident in Fig. 4. It is also apparent that the electron-electron scattering term increases markedly as Ni is added to Pd. The trend of the data in Fig. 4 to higher values as T approaches 0°K is suggestive of a nonnegligible lattice heat conduction, although the scatter of the data caused by the overwhelming importance of the impurity scattering term at the lowest temperatures makes it difficult to definitely establish this trend. The intercepts of the straight lines in Fig. 4 yield values of α , and hence the electron-electron Lorenz number $L_e = A/\alpha$. These values, shown in Table 1, are markedly insensitive to Ni concentration, although they increase somewhat at the higher concentrations.

In the above analysis, we have ignored the possibility of a significant amount of heat conduction via the lattice. At temperatures below about 10°K , it is expected (18) that

Table 1
Parameters of Dilute Pd-Ni Alloys

Ni conc. (nominal) (at-%)	ρ_{297} ($\mu\Omega$ cm)	ρ_0 ($\mu\Omega$ cm)	$10^{12} \times A$ (Ω cm/ $^{\circ}\text{K}^2$)	$10^{15} \times B$ (Ω cm/ $^{\circ}\text{K}^5$)	$10^8 \times L_0^*$ (V^2/K^2)	$10^8 \times L_e^*$ (V^2/K^2)	$10^4 \times D^\dagger$ ($\text{W}/\text{cm } ^{\circ}\text{K}^3$)	$10^4 \times \beta^\dagger$ ($\text{cm}/\text{W}^{\circ}\text{K}$)
(pure Pd)	10.63	0.02686	31.0	11	2.44	1.1	—	2.6
0.21	10.84	0.08866	60.9	11	2.49	1.1	14.0	4.6
0.5	11.08	0.22327	120	4.6	2.48	1.2	4.4	4.8
1.0	11.53	0.36476	273	-54	2.47	1.4	3.0	2.8

*These values are obtained by assuming that lattice heat conduction is negligible.

†These values are obtained by assuming that L_0 is equal to L_s and that $L_e = 1.1 \times 10^{-8} \text{ V}^2/\text{K}^2$ in all specimens. In the case of pure Pd, a value of D could not be meaningfully determined.

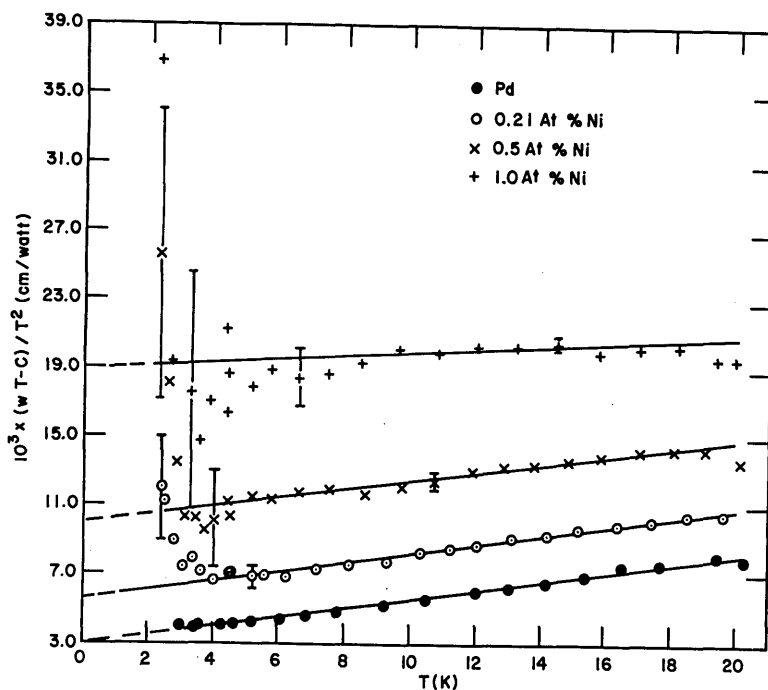


Fig. 4 - Total thermal resistivity w of Pd-Ni alloys as a function of the absolute temperature T . Values of C were obtained by extrapolation to 0°K on a plot of wT vs T^2 . The straight lines have been obtained from least-squares fits of the data to Eq. (13) of the text for Pd and Pd-0.21 at-\% Ni , and were simply drawn through the data for the other specimens. The error bars represent 0.5 percent of the total measured resistivities.

electron scattering of the phonons would dominate, and thus the phonon conduction would exhibit a temperature variation of T^2 . Since conductivities due to different carriers are additive, we can write

$$\frac{1}{w_{e1}} = \frac{1}{w} - DT^2, \quad (14)$$

where w_{e1} is the thermal resistivity appropriate to the electrons alone and D is a constant. The appropriate thermal resistivity for Eq. (13) is of course w_{e1} , and Eq. (13) can thus be modified to the form

$$\left[\frac{T}{(1/w) - DT^2} - C \right] \frac{1}{T^2} = \alpha + \beta T. \quad (15)$$

Inspection of Eq. (15) reveals that the omission of a phonon term tends to reduce the values of α deduced from the data, and hence to increase the apparent values of L_e . This apparent enhancement of L_e would be larger in the more concentrated alloys, where phonon conduction is a larger fraction of the total conduction. Thus without an independent knowledge of the term DT^2 , one cannot obtain reliable values of L_e from the data. We can estimate the magnitude of D for each specimen by using the expression given by Klemens (18),

$$D = 313 \beta^{-1} \theta_D^{-4} N_a^{-4/3}, \quad (16)$$

where θ_D is the Debye temperature and N_a the number of conduction electrons per atom. Since θ_D varies slowly with concentration in this range (23,24), and the value of β for pure Pd is expected to be relatively free of electron-electron effects, the parameters of pure Pd can be used in Eq. (16) to estimate D . We have done this, assuming that $N_a = 0.3$, $\theta_D = 270^\circ\text{K}$, and $\beta = 2.6 \times 10^{-4} \text{ cm/W}^\circ\text{K}$, and obtain $D \approx 10^{-3} \text{ W/cm}^\circ\text{K}^3$. Using this value of D and Eq. (15), we find that the L_e values are not appreciably changed in the pure Pd and Pd-0.21 at-% Ni specimens, although L_e values of the higher concentration alloys now fall below the pure Pd value. However, as pointed out by Fletcher and Greig (25), Eq. (16) tends to overestimate D . Furthermore, in the isoelectronic Pd-Ni alloy system, one would expect D to decrease as the Ni concentration is increased.

We have found that the data for each specimen can be well represented by the following scheme. We determine a value of C by setting $C = \rho_0/L_s$, where L_s is the Sommerfeld value of the Lorenz number, and a value for α by setting $\alpha = A/L_e$, where we take $L_e = 1.1 \times 10^{-8} \text{ V}^2/^\circ\text{K}^2$. With a least-squares technique, we then vary D and β to obtain a best fit of the data for $T \lesssim 10^\circ\text{K}$. The results are presented in Fig. 5, where the straight lines are those obtained from the least-squares calculation. We note that the data are in reasonable accord with this treatment. The values of D obtained in this fashion, as shown in Table 1, are not only the right order of magnitude, but they decrease with increasing Ni concentration. Thus it appears likely that the L_e of Pd is essentially unchanged by the addition of up to 1.0 at-% Ni. However, in this work we take a more conservative interpretation and increase the approximate basic experimental uncertainty of the L_e values of $\pm 10\%$ to an upper limit obtained by ignoring phonon conduction, and a lower limit obtained by assuming a D value of $10^{-3} \text{ W/(cm}^\circ\text{K}^3)$ for all the specimens.

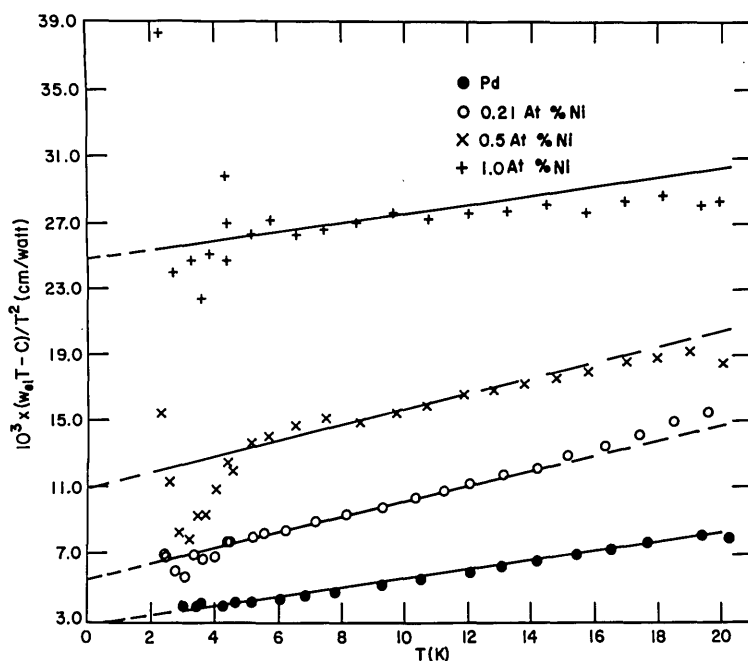


Fig. 5 - Electronic thermal resistivities w_{e1} of Pd-Ni alloys as a function of the absolute temperature T . C was obtained from the electrical data according to $C = \rho_0/L_s$. The straight lines are the results of least-squares fits as discussed in the text.

Thus, in units of $10^{-8} \text{ V}^2/\text{K}^2$, the L_e values are 1.1 ± 0.1 for pure Pd and Pd-0.21 at-% Ni, 1.1 ± 0.2 , -0.5 for Pd-0.5 at-% Ni, and 1.1 ± 0.4 , -0.5 for Pd-1.0 at-% Ni.

COMPARISON OF THEORY AND EXPERIMENT

We turn now to a consideration of specific forms of the integrals $\beta_{(m)}$ defined in the second section which will permit us to compare the measured values of L_e to those values predicted by both the uniform and the local enhancement models. The spectral density function $A(q, \Omega)$ defined above is related to the wave vector and frequency-dependent transverse susceptibility $\chi(q, \Omega)$ by (26)

$$A(q, \Omega) = \frac{1}{i\pi} \text{Im} [\chi(q, \Omega - i\epsilon)] . \quad (17)$$

By using the definition of $\chi(q)$ in Eq. (8) and ignoring the dependence on direction of q , Eq. (17) leads to the following form for $\alpha(q)$ for the case of the uniform enhancement model, with a spherical approximation for the d band:

$$\alpha_U(q) = C_u \frac{(k_s/q) \theta(2k_d - q)}{[1 - I\chi_R^{(0)}(q, 0)]^2} , \quad (18)$$

where C_u is a multiplicative constant whose value does not affect L_e , k_d is the d-electron Fermi wave vector, and $\theta(x) = 0$ when $x < 0$. In Eq. (18), $\chi_R^{(0)}(q, 0)$ is the real part of the dynamic susceptibility of a noninteracting Fermi gas of d electrons and I is the magnitude of the spatially averaged electron-electron Coulomb repulsion of the d electrons in the alloy. Here we have invoked the result of the random phase approximation,

$$\chi(q, \Omega) = \frac{\chi^{(0)}(q, \Omega)}{1 - I\chi^{(0)}(q, \Omega)} .$$

In a similar fashion (9), the expression for $\alpha(q)$ for the local enhancement model can be written as

$$\alpha_L(q) = C_L \left[\frac{\chi_R^{(0)}(q, 0)}{1 - I_{Pd} \chi_R^{(0)}(q, 0)} \right]^2 \quad (19)$$

where I_{Pd} refers to the value of I appropriate to the pure palladium matrix and C_L is again a constant whose value leaves L_e unaffected. The form in Eq. (19) does not depend on any detailed assumption about the shape of the d band. The factor of $1/q$ in Eq. (18) is strictly correct only for the spherically symmetric, parabolic model of the d band. However, one may show that, for a band of arbitrary shape, $\alpha_U(q)$ is proportional to $1/q$ in the small- q limit. Thus, for the general case, Eq. (18) is correct only for small q , but we can assume the form is valid for all values of q and regard it as a rough extrapolation into the large- q region.

It is apparent from Eqs. (18) and (19) and the definitions of $\beta_{(m)}$ that, once the model is chosen, the calculated values of L_e depend only on the form factor $|F(q)|^2$ and on the structure of $\chi_R^{(0)}(q, 0)$. We shall calculate a specific form of $\chi_R^{(0)}(q, 0)$ under the assumption of a spherical d band and introduce a specific form of $|F(q)|^2$. But it is instructive to first consider in a semiquantitative fashion how L_e depends on $\chi_R^{(0)}(q, 0)$ for the two types of enhancement. For this purpose we set $|F(q)|^2 = 1$ and assume that, since the Pd-Ni system under consideration is nearly ferromagnetic, $\chi_R^{(0)}(q, 0)$ has a

maximum value at $q = 0$ and falls off more or less smoothly with increasing q . In order to make some simple numerical estimates, let us assume that $\chi_R^{(0)}(q, 0)$ falls off very slowly with a quadratic dependence on q ; i.e.,

$$\chi_R^{(0)}(q, 0) = N(0) [1 - \zeta q^2],$$

where ζ is an adjustable parameter and $N(0)$ is the unenhanced density of states of the d band evaluated at the Fermi energy. We define $\bar{I} = IN(0)$ and $\xi^2 = \zeta k_s^2$. We recall from the second section that the quantity $Q = \max \{(k_d/k_s), 1\}$ for the spherical model, and assume that $4Q^2 \bar{I} \xi^2$ is small compared to $(1 - \bar{I})$. The integrals $\beta_{(m)}$ are then simply evaluated and, from Eq. (11) we have, for the uniform and local models, respectively,

$$L_e^{(U)} = \frac{5L_s Q^2}{9 + 3Q^2 + \frac{96}{5} \frac{\bar{I} \xi^2 Q^2}{1 - \bar{I}}} \quad (20)$$

and

$$L_e^{(L)} = \frac{5L_s Q^2}{6 + 3Q^2 + 8 \frac{\xi^2 Q^2}{1 - \bar{I}_{Pd}}} \quad (21)$$

In Eqs. (20) and (21) we can see a simple illustration of the effect on the Lorenz numbers of restricting the scattering angle. From the definition of Q , it is apparent that a maximum scattering angle of π yields $Q = 1$, and Q decreases as the scattering is restricted to smaller angles by limiting the value of k_d . Both expressions for L_e become rapidly smaller as Q becomes smaller, i.e., as the scattering becomes restricted to smaller angles. Furthermore, the values of L_e are seen to decrease as ξ increases; the more rapidly $\chi_R^{(0)}(q, 0)$ falls with q , the smaller the Lorenz number. Finally, in Eq. (20), it is apparent that L_e in the case of uniform enhancement falls rapidly as the enhancement factor $1/(1 - \bar{I})$ increases, and thus, in this model, L_e values predicted for Pd-Ni fall rapidly with increasing Ni content.

We see from these simple considerations that the largest values of L_e which can be obtained occur for a $\chi_R^{(0)}(q, 0)$, which is independent of q , and for momentum transfers which permit scattering through all angles. The maximum values are $L_e^{(U)} = 1.018 \times 10^{-8} \text{ V}^2/\text{K}^2$ and $L_e^{(L)} = 1.36 \times 10^{-8} \text{ V}^2/\text{K}^2$, whereas the data for all concentrations yield values of the order of $1.0 \times 10^{-8} \text{ V}^2/\text{K}^2$. It will be shown that in a more detailed treatment we will obtain agreement with the data by assuming $\chi_R^{(0)}(q, 0)$ falls off slowly with q , with Q near unity.

It should be pointed out that the value of $L_e^{(L)}$ obtained from Eq. (21) with $Q = 1$ and $\xi = 0$ is the value of the Lorenz number appropriate to electron-electron scattering when all angular dependence of the scattering matrix element is neglected. In other words, in this limit, our calculation reduces to a calculation of the Lorenz number for spherically symmetric scattering by single-particle excitations of low frequency which dominate the scattering at low temperatures. This is the Lorenz number calculated by Herring (13), who postulated that the complexities of the Fermi surfaces in the transition metals would result in a lack of angular dependence of the scattering. In fact, Herring's value of $1.58 \times 10^{-8} \text{ V}^2/\text{K}^2$, which was calculated in the absence of impurity scattering, reduces to a modified value (27) of about $1.35 \times 10^{-8} \text{ V}^2/\text{K}^2$ when the effect of the presence of very strong impurity scattering is included. This is just the value we obtained above by using for our variational calculation, trial functions appropriate to the presence of a large amount of impurity scattering.

In an attempt to fit the data of the third section we have assumed that d electrons occupy a simple spherical band. In this case the real part of the dynamic susceptibility can be written as

$$\chi_R^{(0)}(q, 0) = N(0) \left[\frac{1}{2} h \left(\frac{q}{2k_d} \right) \right], \quad (22)$$

where $h(x)$ is the Lindhard function

$$h(x) = 1 + \frac{1 - x^2}{2x} \ln \left| \frac{1 + x}{1 - x} \right|. \quad (23)$$

Then the integrals $\beta(m)$ become

$$\beta^{(U)}(m) = \int_0^{2Q} d\eta \eta^{m-1} \left\{ 1 - \bar{I} \left[\frac{1}{2} h \left(\frac{k_s \eta}{2k_d} \right) \right] \right\}^{-2} |F(k_s \eta)|^2 \quad (24)$$

and

$$\beta^{(L)}(m) = \int_0^{2Q} d\eta \eta^m \left\{ \frac{N(0) \left[\frac{1}{2} h \left(\frac{k_s \eta}{2k_d} \right) \right]}{1 - \bar{I}_{Pd} \left[\frac{1}{2} h \left(\frac{k_s \eta}{2k_d} \right) \right]} \right\}^2 |F(k_s \eta)|^2. \quad (25)$$

To calculate explicit values of L_e from Eqs. (24) and (25), it is necessary to adopt values for the parameters as well as an explicit expression for $F(k_s \eta)$. A value of k_s was obtained by using the de Haas-van Alphen data of Vuillemin (28) for Pd and approximating the roughly spherical s -like portion of the Fermi surface with a sphere of radius $k_s = 9.125 \times 10^7 \text{ cm}^{-1}$. We treat k_d as an adjustable parameter, while for \bar{I}_{Pd} we assume the value of 0.9.* Since the d -like portion of the Pd Fermi surface is very complex, we regard k_d as a parameter which characterizes in an average way the manner in which $\chi_R^{(0)}(q, 0)$ falls off with increasing q . We shall not attempt to associate k_d with any feature of the grossly nonspherical d -hole surfaces in Pd.

$F(q)$ was obtained by using the radial portion of the $4d$ (10) orbitals of atomic Pd calculated by Herman and Skilman (29). It was found that the dominant features of the square of the tabulated radial wave function could be roughly represented by a function of the form $r^2 \exp(-r^2/r_0^2)$ with an r_0 of $0.58 \times 10^{-8} \text{ cm}$. The Fourier transform of this function, normalized to unity at $q = 0$, yields for $F(q)$:

$$F(q) = [1 - 1/6 (qr_0)^2] \exp[-(qr_0)^2/4]. \quad (26)$$

It is interesting that the Lorenz numbers are fairly sensitive to the form of $F(q)$. The form shown in Eq. (26) yields a Lorenz number which is reduced by roughly 20% below the value obtained with an $|F(q)|^2$ of unity for all values of q . Other functions which we have tried yield an even larger reduction of L_e . A representation of the d orbital by $|\Psi(r)|^2 \approx \exp(-r/r_0)$, for example, reduces the Lorenz number by more than a factor of two.

*The value of \bar{I}_{Pd} has not been definitely established, but 0.9 is the generally accepted value (cf. S. Foner and E. J. McNiff, Jr., Phys. Rev. Letters 19:1438 (1967)).

To compare the calculated Lorenz numbers to the data, we define the parameter $\delta A/A_{Pd}$ by

$$\frac{\delta A}{A_{Pd}} = \frac{A_{alloy} - A_{Pd}}{A_{Pd}}, \quad (27)$$

where the electrical resistivity due to electron-electron scattering is represented by AT^2 . Consider first the uniform enhancement model. For a given choice of k_d , Eq. (24) can be used together with Eqs. (10) and (11) to calculate both ρ_e and L_e as functions of \bar{I} . Then by noting that for pure Pd, $\bar{I} \rightarrow \bar{I}_{Pd}$, and by using the definition in Eq. (27), the parameter \bar{I} can be eliminated to yield $L_e^{(U)}$ as a function of $\delta A/A_{Pd}$.

For the local enhancement model, Eqs. (25) and (11) yield a Lorenz number appropriate to the scattering at the nickel sites only. It is necessary to account for the scattering by the host Pd matrix, which exhibits a Lorenz number obtained by the uniform enhancement model with \bar{I} equal to \bar{I}_{Pd} . If c is the concentration of Ni atoms, for small concentrations one can write

$$L_e = \frac{\rho_e^{(Pd)} + c\rho_e^{(Ni)}}{[w_e^{(Pd)} + cw_e^{(Ni)}]T},$$

which, after a little algebra, becomes

$$L_e = L_{Pd} \frac{1 + \frac{\delta A}{A_{Pd}}}{1 + \frac{L_{Pd}}{L_{Ni}} \frac{\delta A}{A_{Pd}}}, \quad (28)$$

where L_{Pd} is the pure Pd value of L_e , and $L_{Ni} = (\rho_e^{(Ni)}/w_e^{(Ni)}T)$. In applying Eq. (28) we identify L_{Pd} with an L_e calculated from the uniform enhancement model with $\bar{I} = \bar{I}_{Pd}$, while L_{Ni} is the L_e calculated with the local enhancement model.

The integrals $\beta(m)$ were evaluated by standard computer techniques, and the results are shown in Fig. 6 as a plot of L_e as a function of $\delta A/A_{Pd}$. Calculations were carried out for various values of k_d , and it was found that the computed values of L_e were an order of magnitude smaller than the measured values unless k_d was set larger than k_s . The theoretical curves exhibited in Fig. 6 were obtained with $k_d = 2k_s$. Note that the uniform enhancement model yields an L_e which falls off much too rapidly with increasing Ni content. This is caused, as we have seen, by this model's strong increase of small-angle scattering with increasing enhancement.

The largest discrepancy between theory and experiment is seen to be at pure Pd. A natural question is whether or not this discrepancy is caused by the omission of Baber scattering in our treatment of the resistivities of the pure matrix. To investigate this possibility, we have calculated the contribution of Baber scattering to the resistivities of these materials. The matrix element for screened Coulomb interactions is proportional to $1/(q^2 + \kappa^2)$, where κ is the Fermi-Thomas screening length. Now κ^2 is given by $4\pi e^2 N(0)$, where $N(0)$ is the total density of states at the Fermi level. In materials with unfilled d bands, $N(0)$ is dominated by the d-band contribution rather than the s-band, as Rice has assumed (11). To obtain an estimate of κ , one notes that for s electrons in a spherical band, $(4\pi e^2 N(0)_s)^{1/2}$ would give a $\kappa_s \approx 15 \times 10^7 \text{ cm}^{-1}$. Since the density of states of the d electrons is known to be much larger than that of the s electrons, κ will be much larger than κ_s , and $\kappa \gg q$ over the entire range of q . Thus the matrix element can be replaced by a constant. We then have, apart from the multiplicative factor,

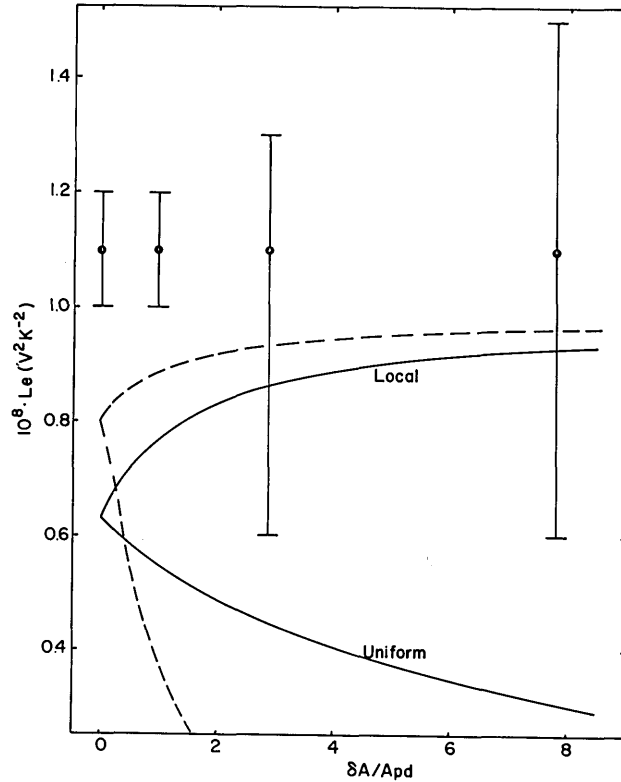


Fig. 6 - Comparison of calculated and measured values of L_e for Pd and dilute Pd-Ni alloys. A is the coefficient of the T^2 term in the electrical resistivity, and $\delta A = A_{\text{alloy}} - A_{\text{Pd}}$. The solid dots and error bars represent values of L_e extracted from the data. The solid lines are the results of calculations for both local and uniform enhancement models of spin density fluctuation scattering with $\bar{I}_{\text{Pd}} = 0.9$, $k_s = 9.125 \times 10^7 \text{ cm}^{-1}$, and $k_d = 2k_s$. The dashed lines are the results of modifying the calculation by adding a concentration-independent amount of Baber scattering to the spin density fluctuation scattering; the dashed curves shown here assume 90 percent of the scattering in pure Pd is Baber scattering.

$$\beta^{(B)}(m) = \int_0^{2Q} d\eta \eta^{m-1} |F(k_s \eta)|^2. \quad (29)$$

For pure Baber scattering, with the $F(q)$ and value of k_s used above, Eqs. (29) and (11) yield a Lorenz number of $0.8304 \times 10^{-8} \text{ V}^2/\text{K}^2$. Therefore, even if one assumed that all of the electron-electron scattering in pure Pd was due to Baber scattering, the calculated Lorenz number would not agree with the experimental result. By assuming that Baber scattering and electron-paramagnon scattering contribute additively to the resistivities and that the same amount of Baber scattering present in pure Pd is also present in the alloys, the curves in Fig. 6 can be modified to include Baber scattering.

The modified curves shown in Fig. 6 indicate the effect of assuming that 90 percent of the scattering in pure Pd is due to screened Coulomb interactions.

DISCUSSION

The most striking feature of this work is that it is clear that a $\chi_R^{(0)}(q, 0)$ which varies slowly in the range $0 < q < 2k_s$ is required to represent the data. Although our most detailed calculations were carried out with the somewhat unrealistic model of a spherical d band, it is apparent from arguments presented in the previous section that only a rather flat $\chi_R^{(0)}(q, 0)$ function for these values of q is consistent with the data. We note, however, that recently Kim and Schwartz (30) concluded from their interpretation of Low and Holden's (31) neutron scattering data on dilute Pd-Fe alloys that the q dependence of the susceptibility function is significantly stronger than that predicted for a parabolic band of d electrons with k_d chosen to fit the number of d holes present in Pd. Using the small- q approximation to the Lindhard function, the susceptibility for a parabolic band of d electrons is proportional to $[1 - (1/12)(q/k_d)^2]$. Kim and Schwartz find that, with k_d adjusted to $5 \times 10^7 \text{ cm}^{-1}$, this dependence on q is not strong enough to fit their model to the data of Low and Holden, whereas we find that the much weaker falloff obtained with $k_d \approx 18 \times 10^7 \text{ cm}^{-1}$ is required to fit our data. It should be noted that the theory of Kim and Schwartz assumes that the moment in the Fe cell perturbs the surrounding Pd matrix in a fashion describable by linear response theory. This linear response theory predicts that the total moment varies directly as the susceptibility of the host matrix. That these moments do, indeed, saturate is well known. For example, Geballe and coworkers (32) found in their measurements of the giant moment of Fe atoms in Ir-Pt, Pt-Pd, and Rh-Pd matrices that, as a function of the susceptibility of the matrix, the moment per Fe atom falls on a single curve which shows saturation effects at about 8 Bohr magnetons. Since the moment of Fe in palladium is of the order of 10 or 12 Bohr magnetons, the use of a linear theory may be a poor approximation.

On the other hand, the local-enhancement model employed in the present work gives a good correlation between the measured values of the coefficients of the electron-electron electrical resistivities and the magnetic susceptibilities of the dilute Pd-Ni alloys. Although the presence of a phonon heat conduction causes some uncertainty in the analysis of our data, it is clear that a very slowly varying $\chi_R^{(0)}(q, 0)$ for $0 < q < 2k_s$ is required to even approach the measured values of the Lorenz number. Furthermore, this result does not appear inconsistent with the preliminary results of a band-structure calculation of the frequency and wave-vector dependence of $\chi_R^{(0)}$ which is being carried out by Missetich (33). We feel, therefore, that a slowly varying $\chi_R^{(0)}(q, 0)$ is more appropriate to Pd, although a definite conclusion must await further developments.

As pointed out above, Herring has postulated that the same value of the electron-electron scattering Lorenz number $1.35 \times 10^{-8} \text{ V}^2/\text{°K}^2$ may be appropriate to all the transition metals due to their common property of exhibiting complex Fermi surfaces. His value is somewhat larger than the data presented here, at least for the Pd and Pd 0.21 at-% Ni specimens, although in view of the simplicity of his model, it is reasonably close to the measured value. On the other hand, in the course of the present work we have found that the calculated values of the Lorenz number are quite sensitive, not only to the angular dependence of the scattering, but also to the details of the shape of the Wannier function associated with the d band. We note, also, that in the present work we obtained Lorenz number values which were lower than the experimental values at the dilute end of the concentration range, although the agreement between theory and experiment was somewhat better at the higher concentrations. We attribute this to our omission of any detailed band-structure effects which would be more important for pure Pd and the more dilute alloys, where the scattering by the matrix contributes a larger fraction to the total scattering than does the scattering at the Ni sites. Since the details just

outlined seem to be important for calculating Lorenz number values appropriate to Pd and Pd-Ni, we feel that such details may also be important in other transition metals.

To the extent that a two-band model of the type employed here is applicable to other transition metals, these calculations provide an estimate of the electron-electron scattering Lorenz number for Baber scattering. Since the interaction is screened by the *d* electrons, as noted above, and provided that scattering to all angles is possible, Eqs. (29) and (11) show that the Lorenz number depends only on the parameter k_s . In fact, in the absence of a form factor, the Baber scattering Lorenz number predicted here would be the same in all transition metals and would have a value of $1.02 \times 10^{-8} \text{ V}^2/\text{°K}^2$. However, as we saw above, the form factor is important; in Pd the form factor reduces the Baber scattering Lorenz number by roughly 20 percent.

The experimental value of L_e for rhenium has been found to be $L_e \approx 0.9 \times 10^{-8} \text{ V}^2/\text{°K}^2$ (4),* and it is interesting to compare this value to an estimate obtained from the present Baber scattering model. For this purpose we employ the model of the Fermi surface of hcp rhenium suggested by Mattheiss (34). Although the Fermi surface of Re is rather elaborate, a crude representation can be made. A central, roughly cylindrical electron sheet centered about the Γ -A axis contains about 0.16 electrons per atom, while roughly spherical hole surfaces, centered about the points L on the surface of the Brillouin zone, contain about 0.15 holes per atom. Other parts of the Fermi surface contain numbers of electrons per atom which are an order of magnitude smaller than these values. We can very crudely apply the two-band model by using a sphere of radius roughly equal to the central cylinder's radius to represent the *s* electrons, and by assuming that the dominant scattering of the *s* electrons is due to the *d* electrons responsible for the hole surfaces of Re. The value of k_s is then roughly $5.3 \times 10^7/\text{cm}^{-1}$. We apply the same function for $F(q)$ as we used for Pd, but find that a value of r_0 of $0.68 \times 10^{-8} \text{ cm}$ is appropriate for the 5*d* (5) orbitals of Re as tabulated by Herman and Skilman (29). We then obtain from Eqs. (29) and (11) a value of $L_e = 0.93 \times 10^{-8} \text{ V}^2/\text{°K}^2$. The model of Re we have used is, indeed, crude, particularly since the model is spherically symmetric while some anisotropy has been observed in the resistivities. Thus the excellent agreement we obtained is somewhat fortuitous. Nevertheless, we feel this agreement suggests that our model contains the essential features of Baber scattering, namely that the screening is accomplished by the *d* electrons and that the form factors of the Wannier functions in the *d* band are important in determining the numerical value of L_e .

ACKNOWLEDGMENTS

It is a pleasure to acknowledge the technical advice of B. C. La Roy and the assistance of R. Williams in alloy preparation. We are indebted to O. R. Gates and E. J. Brooks of the Analytical Chemistry Branch, NRL, for performing the chemical characterizations of our materials. We also wish to acknowledge Dr. M. J. Rice for interesting discussions, and we are grateful to Dr. A. M. Missetich for his helpful comments. One of the authors, Dr. J. T. Schriempf, wishes to thank Dr. A. C. Ehrlich for many stimulating discussions, and also to convey his appreciation to the Physics Department, University of California at Irvine, for their kind hospitality during the final stages of this work.

This work was supported in part by the Air Force Office of Scientific Research, Office of Aerospace Research, USAF, under AFOSR grant number 68-1448.

*The experimental value of L_e for Re originally reported in Ref. 4 was in error. This is discussed in Ref. 6.

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Appendix

EVALUATION OF INTEGRALS I_a , I_b , AND I_c

We present here the method of converting the integrals I_a , I_b , and I_c to forms which are listed in integral tables.* Full details will be given for I_b ; I_a and I_c can be evaluated by the same technique.

First replace $x - x'$ by y , and x by $y + x'$. The integral I_b defined in the second section becomes

$$I_b = \int_{-\infty}^{\infty} dy dx' (y^2 x' + x'^2 y) f^0(y + x') [1 - f^0(x')] [1 + n(y)] ,$$

or, upon replacing the variable of integration x' by x and introducing the explicit forms of $f^0(x)$ and $n(x)$,

$$I_b = \int_{-\infty}^{\infty} \frac{(x^2 y + y^2 x) dx dy}{(1 + e^{-x})(1 - e^{-y})(1 + e^{y+x})} \quad (A1)$$

We introduce the integrals I_n and J_m such that

$$I_b = I_1 + I_2 , \quad (A2)$$

$$I_n = \int_{-\infty}^{\infty} \frac{J_{3-n} x^n dx}{(1 + e^{-x})} , \quad n = 1, 2 , \quad (A3)$$

$$J_m = \int_{-\infty}^{\infty} \frac{y^m dy}{(1 - e^{-y})(1 + e^{y+x})} \quad m = 1, 2 . \quad (A4)$$

The integrals J_m can be reduced to tabulated forms by the substitution $\mu = e^y$. One obtains

$$J_m = e^{-x} \int_0^{\infty} \frac{(\ln \mu)^m d\mu}{(\mu - 1)(\mu + e^{-x})} . \quad (A5)$$

Using Gradshteyn and Ryzhik's (35) equations 4.232.3 and 4.261.4,

$$J_m = \frac{(\pi^2 + x^2)(-x)^{m-1}}{(1 + e^{-x})(m+1)} , \quad m = 1, 2 , \quad (A6)$$

With Eq. (A6), the expression (A3) can now be written as

*This technique was suggested to us by J. Oitmaa and A. Grimm.

$$I_n = \frac{(-1)^{2-n}}{4-n} \int_{-\infty}^{\infty} \frac{(\pi^2 + x^2) x^2 dx}{(1 + e^{-x})^2}, \quad n = 1, 2. \quad (A7)$$

By employing the hyperbolic cosine and Gradshteyn and Ryzhik's formula 3.527.5 the integral in Eq. (A7) is readily evaluated:

$$I_n = \frac{(-1)^{2-n}}{4-n} \left(\frac{4}{5} \pi^4 \right). \quad (A8)$$

Hence, from Eqs. (A2) and (A8) the final value of I_b is seen to be $2\pi^4/15$.

DOCUMENT CONTROL DATA - R & D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY (Corporate author) Naval Research Laboratory Washington, D.C. 20390		2a. REPORT SECURITY CLASSIFICATION Unclassified	
		2b. GROUP	
3. REPORT TITLE ELECTRON-ELECTRON SCATTERING EFFECTS IN THE LOW-TEMPERATURE LORENZ NUMBERS OF DILUTE <u>Pd-Ni</u> ALLOYS			
4. DESCRIPTIVE NOTES (Type of report and inclusive dates) ... Completes one phase of problem; work continues on other aspects.			
5. AUTHOR(S) (First name, middle initial, last name) J.T. Schriempf, A.I. Schindler, D.L. Mills			
6. REPORT DATE September 15, 1969		7a. TOTAL NO. OF PAGES 32	7b. NO. OF REFS 35
8a. CONTRACT OR GRANT NO. NRL Problem No. M01-10		9a. ORIGINATOR'S REPORT NUMBER(S) NRL Report 6949	
b. PROJECT NO. RR 007-01-46-5408		9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report)	
c.			
d.			
10. DISTRIBUTION STATEMENT This document has been approved for public release and sale; its distribution is unlimited.			
11. SUPPLEMENTARY NOTES		12. SPONSORING MILITARY ACTIVITY Department of the Navy (Office of Naval Research), Washington, D.C. 20360	
13. ABSTRACT The thermal and electrical resistivities of <u>Pd-Ni</u> alloys at concentrations ranging from 0 to 1.0 at-% Ni have been measured at temperatures between 2 and 20°K. The Lorenz numbers L_e appropriate to electron-electron scattering have been extracted from the data and are found to be relatively insensitive to Ni concentration. A theoretical calculation of L_e has been carried out under the assumption that the entire current is carried by s electrons which are scattered from spin density fluctuations of the electrons in the d band. A consistent interpretation of the data is obtained by employing the local enhancement model, while the uniform enhancement model is found to yield values of L_e which decrease too rapidly with increasing Ni concentration to reasonably represent the experimental results. The calculated value of L_e is significantly smaller than the experimental value for pure Pd; it is felt this may be due to the neglect of detailed band structure effects in this work. Lorenz numbers for Baber scattering have also been calculated by using Coulomb interaction screened by the d electrons. It is found that the presence of Baber scattering cannot remove the discrepancy between theory and experiment for pure Pd. The Baber scattering model is also used to calculate the L_e for rhenium and gives very good agreement with experiment.			

14. KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Thermal conductivity Electrical resistivity Lorenz number Electron-electron interactions Transport properties Palladium alloys						